

# Theoretical Investigation of the Magnetic Order in FeAs

Dobysheva Lyudmila V. <sup>a</sup> and Arzhnikov Anatoly K. <sup>b</sup>

Phys.-Techn. Institute UrBr Russian Ac. Sci., 426001, Izhevsk, Kirova str., 132, RUSSIA

<sup>a</sup>lyu@otf.pti.udm.ru, <sup>b</sup>arzhnikov@otf.pti.udm.ru

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**Abstract.** The magnetic structure of the iron monoarsenide FeAs is studied using first-principles calculations. We consider the collinear and non-collinear (spin-spiral wave) magnetic ordering and magnetic anisotropy. It is analitically shown that a magnetic triaxial anisotropy results in a sum of two spin-spiral waves with opposite directions of wave vectors and different spin amplitudes, so that the magnetic moments in two perpendicular directions do not equal each other.

## Introduction

The discovery in 2008 of a new class of superconductors, namely, layered compounds on the basis of iron [1] attracted a great interest. This broke the monopoly of cuprates in the high-temperature-superconductivity (HTSC) physics and aroused an expectance for the progress in theoretical understanding of the HTSC. The new class of superconductors demonstrated both common and different features in comparison to cuprates [2]. One of the common points is a presence of a strong magnetisation. The majority of researchers believe that, in these superconductors, magnetic fluctuations are responsible for the electron pairing as in the case of cuprates. That is why numerous works, including the presented one, are devoted to investigation of the magnetic properties.

The FeAs attracts large interest due to extraordinary properties of its close relatives such as LaFeAsO, BaFe<sub>2</sub>As<sub>2</sub>, and NaFeAs, which are attributed to the presence of FeAs planes with the same local structure as in the iron monoarsenide. The experimental work conducted in 2011 [3] using a polarised neutrons revealed a row of peculiarities which are important for theory. In particular, direct measurements of spin-spiral waves that are the ground state of the system were conducted. However, the authors interpretation in that work based on the fitting of the parameters of a Heisenberg Hamiltonian is misleading, as d-electrons which form the magnetic moment are delocalised, and correlations are weak [2].

In connexion with this, here, a first-principles calculation of magnetic properties of FeAs crystal is conducted and an interpretation of the anisotropy of the magnetic moment of spin-spiral wave is given in the frames of a model Hamiltonian.

## First-principles calculation of the electron structure

The crystal structure of FeAs is well known from experiments, for example, [4]. It forms the orthorhombic (Pnma) MnP-type crystal (Fig. 1).

The calculations are conducted with a full-potential linearized augmented waves (FP-LAPW) method realized in the package WIEN2k [5]. In this method the unit cell is divided into two parts: the nonoverlapping atomic spheres and the interstitial region. In the latter the wave functions are expanded into plane waves; inside the spheres the plane waves are augmented

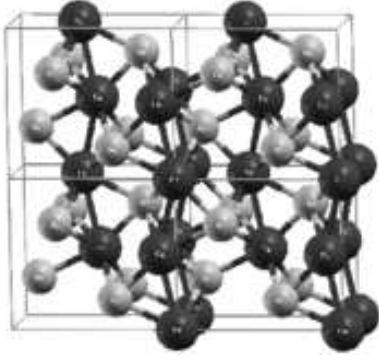


Fig. 1: 2x2 unit cell of FeAs.

by an atomic-like spherical harmonics expansion with radial wave functions. The exchange - correlation potential is calculated using the "local density approximation" (LDA) [7] and the "generalized gradient approximation" (GGA) [6]. All parameters of the calculation scheme are chosen in a standard for the method way. The basic results are tested for numerical convergence with respect to the calculation parameters, they do not also depend on the approximation of the exchange-correlation potential: GGA [6] or LDA [7].

With the experimental lattice parameters [4]  $a = 0.60278$  nm,  $b = 0.54420$  nm, and  $c = 0.33727$  nm, the precise positions of atoms in the unit cell that correspond to the minimum energy are found through optimisation of internal parameters in the GGA potential. The optimized positions are as follows:

Fe1 = (0.0, 0.0, 0.0), Fe2 = (0.39895, 0.99961, 0.5),  
 Fe3 = (0.89895, 0.5, 0.0), Fe4 = (0.5, 0.49961, 0.5),  
 As1 = (0.62404, 0.19863, 0.0), As2 = (0.77491, 0.80098, 0.5),  
 As3 = (0.27491, 0.69863, 0.0), As4 = (0.12404, 0.30098, 0.5),

which are close to the experimentally found positions.

Further, using the experimental lattice parameters and the optimised atomic positions, considered are four magnetic states with collinear magnetic moments. In one of them all Fe local magnetic moments have the same direction (ferromagnetic FM order), the other three have both positive and negative moments with zero overall magnetization and are denominated as AFM1 (Fe1 up, Fe2 up, Fe3 down, Fe4 down), AFM2 (Fe1 up, Fe2 down, Fe3 up, Fe4 down), and AFM3 (Fe1 up, Fe2 down, Fe3 down, Fe4 up). Different collinear solutions (FM and AFMs) are obtained using different starting potentials.

The AFM solutions are slightly lower in energy than the FM state (Table 1). The state

Table 1: The energy  $E$  and atomic magnetic moments  $M$  in four collinear magnetic states considered in FeAs

magnetic state	$E(\text{Ry})$	$M$
FM	-28271.25146	0.62
AFM1 uudd	-28271.25916	1.00
AFM2 udud	-28271.26015	0.99
AFM3 uddu	-28271.26471	1.23

AFM3 is lowest by energy, the FM state being higher by 13 mRy. A calculation with account of the spin-orbit interaction is conducted and the magnetocrystalline anisotropy is obtained. The magnetocrystalline anisotropy obtained as a difference of total energy between the states with magnetisation along the directions specified is equal to  $E_{010} - E_{001} = 1\mu\text{Ry}$  and  $E_{100} - E_{001} = 36\mu\text{Ry}$ .

The optimization of the lattice parameters in the LDA exchange-correlation potential confirms a well-known tendency: the GGA potential allows to obtain structural parameters closer to experiment, whereas the LDA potential gives the parameters further from the experimental ones but describes better than the GGA potential magnetic moment which is  $M_{LDA} = 0.65\mu_B$  for the AFM3 state (compare to experiment in [3]  $M = 0.58 \pm 0.06\mu_B$ ).

The possibility of a few magnetic states of a system means that an intermediate state is the state with a minimum energy. We conducted a calculation of energy of spin-spiral wave depending on its wave vector  $q$ . The calculation is performed with a non-collinear-magnetic version of WIEN2k package [8, 9].

$E(q)$  curves obtained show that the most energetically favourable state AFM3 has a minimum energy at  $q = 0$  (the directions 100 and 001 of  $q$  were considered). In the FM state, the

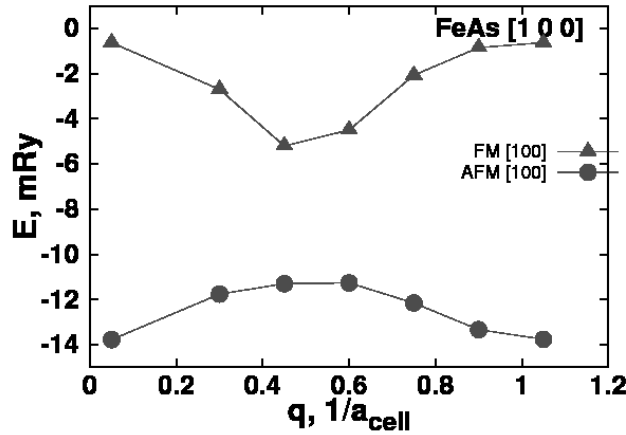


Fig. 2: Energy dependence of the spin-spiral wave on the  $q$  wave vector in FeAs.

SSW with the wave vector  $q = 0.5a^{-1}$  in 100 direction possesses the minimum energy (here  $a$  is the lattice parameter), the SSW with the 001 direction of  $q$  having a minimum at  $q = 0$ . The difference between the collinear and the SSW solution at minimum energy is about 5 mRy/cell, see fig.2.

We have also studied the solutions at the lattice parameters obtained in the LDA of exchange-correlation potential after minimization by energy. Qualitatively the results repeat, though the numerical differences between the states are smaller.

## Anisotropy

The calculations of the state with minimum energy, that is the AFM3 state, show that the system has a triaxial anisotropy, the energy of the state with magnetisation perpendicular to the iron planes (that is  $M \parallel 100$ ) being significantly higher than in cases with  $M \parallel 001$  or  $010$ . So, the magnetic moment should lie in the plane of iron atoms, which is confirmed experimentally in [3]. At the same time, there is a slight magnetic anisotropy in the plane between the directions 010 and 001 (the difference of 1  $\mu$ Ry is on the level of calculational errors). Therefore we assume that magnetic anisotropy of the FeAs compound can be described by an angle between an easy axis  $x$  in the iron plane which we denote here as  $xy$ , or by the projection of the magnetic moment to the  $x$  axis:

$$E_{anis} = - \sum_i \sum_i 4D(S_i^{(x)})^2 \quad (1)$$

Adding this contributon to the Hubbard Hamiltonian, we obtain:

$$H = \frac{1}{N} \sum_{k,\sigma} \epsilon_k^0 c_{k\sigma}^+ c_{k\sigma} - \frac{U}{2N} \sum_j c_{j\uparrow}^+ c_{j\uparrow} c_{j\downarrow}^+ c_{j\downarrow} - \sum_j D (c_{j\uparrow}^+ c_{j\downarrow} + c_{j\downarrow}^+ c_{j\uparrow})^2 \quad (2)$$

Here we use the equality

$$2S_j^x = (S_j^+ + S_j^-) = (c_{j\uparrow}^+ c_{j\downarrow} + c_{j\downarrow}^+ c_{j\uparrow}) \quad (3)$$

The simplest approximation of this Hamiltonian over the class of the wave functions of the spin-spiral waves is a mean field approximation (MFA). Then the Hamiltonian of the system with anisotropy can be rewritten as:

$$\begin{aligned} H = & (U + 3D)(M_1^2 + M_2^2) + Un_{el}/2 + 1/N \sum_{k,\sigma} \epsilon_k^0 c_{k\sigma}^+ c_{k\sigma} \\ & + [-(U + 2D)M_1 - DM_2] \frac{1}{N} \sum_Q (S_Q^+ + S_Q^-) \\ & + [-(U + 2D)M_2 - DM_1] \frac{1}{N} \sum_Q (S_{-Q}^+ + S_{-Q}^-) \end{aligned} \quad (4)$$

where  $S_Q^+ = \sum_k c_{k\uparrow}^+ c_{k+Q\downarrow}$ ,  $S_Q^- = \sum_k c_{k\uparrow}^+ c_{k-Q\downarrow}$ ,  $\langle S_Q^+ \rangle = \langle S_Q^- \rangle = M_1$ ,  $\langle S_{-Q}^+ \rangle = \langle S_{-Q}^- \rangle = M_2$ .

Solution of the system is a superposition of two spin-spiral wave of opposite directions with a total magnetisation:

$$\vec{M} = \vec{e}_x (M_1 + M_2) \cos \vec{Q} \vec{R}_j + \vec{e}_y (M_1 - M_2) \sin \vec{Q} \vec{R}_j \quad (5)$$

Different projections of the magnetization on the axis x and y were actually revealed in experiment [3].

## Summary

Using an ab-initio method of the electron structure calculation (FP LAPW that is realized in WIEN2k package), we have studied the magnetic structure of FeAs, both GGA and LDA potentials being used in calculations. After finding the optimum atomic positions in the unit cell, we have considered a few magnetic configurations with a collinear structure. There are a ferromagnetic FM and three kinds of antiferromagnetic AFM structures found. The AFM structures have lower total energies than the FM one. For all four structures, the calculations with spin-orbit term included have been conducted and the magnetic anisotropy has been studied.

Using a package version for the noncollinear magnetism, the dependence of the total energy on the wave vector q has been obtained.

With a model Hamiltonian, it has been shown that triaxial magnetic anisotropy results in a spin-spiral waves with a corresponding difference in the spin amplitudes, so that the magnetic moments in two perpendicular directions do not equal each other, and the spin spiral becomes elliptic.

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